

PROPERTIES OF METALLIC FILMS IN PRECISE CALCULATION OF THE CASIMIR FORCE

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Optical properties of the deposited gold films are discussed in connection with the Casimir force prediction. Voids in the films and electron scattering on the grain boundaries reduce the force at small separations on the 2 % level in comparison with the bulk material prediction. The contribution of the patch potential due to polycrystalline structure of the films is shown to be small for the existing Casimir force experiments.

1 Introduction

Detailed investigation of the Casimir force¹ between two uncharged metallic plates, predicted in 1948, became possible only recently with the progress in microtechnologies allowing to control separation between bodies smaller than $1 \mu m$. The development of the experiment^{2,3,4,5} is characterized by steady improvement in the precision of the force measurement from 5 %² to 0.25 %⁵. The progress in theoretical prediction of the force is not so impressive. Starting from the first precise calculations^{6,7,8,9} and up to now⁵ the force is calculated using the optical properties of bulk metals taken in the handbooks. However, in all the experiments the force was measured between deposited metallic films. Typically it was gold with the thickness $100 - 200 nm$. It is clear that the deposited material and bulk material are different if we pretend on high precision calculations. In this paper the question is analyzed to what extend the properties of real deposited metallic films can change the force in comparison with that predicted for the bulk material. To answer this question we inevitably have to turn to the material science. The steps in this direction never been done before but without understanding of the used material there will be no further progress in the prediction of the Casimir force.

Originally the Casimir force¹ was calculated between ideal metallic plates. In this case it does not include any material parameters: $F_c = (\pi^2/240) (\hbar c/a^4)$, where a is the separation between plates. At small separations the deviation of the used metal from the ideal one becomes significant and one has to calculate the force using a more general Lifshitz formula¹⁰. This formula takes into account real optical properties of the plate material via the dielectric function $\varepsilon(\omega)$, which is taken in the handbooks¹¹. Gold is the best material for the force measurement since it is chemically inactive and its low frequency behavior, where the handbook data unaccessible, can

be reliably predicted with the Drude model.

2 Voids in the films

Optical properties of *Au* films were investigated widely in 60th-70th. The results are collected in the handbooks¹¹. Significant deviation in the optical data was attributed to genuine sample differences caused by different sample preparation methods¹¹. The handbook data¹¹ were carefully chosen to represent the bulk material as close as possible. A special investigation of the sample preparation effect was undertaken¹² with the conclusion that the most significant role played voids in the samples. Annealed films demonstrated larger density due to larger grain size. A single parameter model representing voids in an otherwise homogeneous medium was shown to account for the major discrepancies in the above-band-gap ($E = 2.5$ eV) spectra for *Au* samples prepared in different ways. It allowed to get information about the volume fraction of voids f_V in different samples. For $f_V \ll 1$ and $|\varepsilon| \gg 1$ the model gave the effective dielectric function $\langle \varepsilon \rangle$ defined by the relation $\langle \varepsilon \rangle = \varepsilon (1 - \frac{3}{2}f_V)$. For the film (150 nm) evaporated with e-beam on *NaCl* substrate at room temperature with a deposition rate of 23 Å/s the volume of voids was found to be larger on 4 % than in the annealed samples used for the handbook data¹¹. For the 50 nm thick sputtered films deposited at the rate smaller than 13 Å/s the volume of voids was found to be 4 % at room temperature deposition and 1 % at 250° C. The cleaved *NaCl* substrate was chosen because the films start to grow epitaxially on it. The same conclusion was made in a more recent investigation¹³.

In the most sensitive experiments the *Au* films were evaporated³ or sputtered⁵ at room temperature, no annealing was reported. One can expect¹² that in both cases the volume of voids was of about of 4 %.

The correction to the Casimir force due presence of voids was calculated using the data¹¹ for ε . These data were scaled in accordance with the effective dielectric function $\langle \varepsilon \rangle = \varepsilon (1 - \frac{3}{2}f_V)$. Calculations were performed in the way similar to⁷ for plate-plate and sphere-plate geometries. The results for the relative correction to the force are shown in Fig. 1(a) for minimal separations in the experiments: $a = 63$ nm for sphere-plate³ and $a = 260$ nm for the effective plate-plate geometries⁵. Let us stress that this correction always make the force for films smaller than that for the bulk material.

3 Scattering on the grain boundaries

It is well known that the resistivity of deposited films deviates significantly from the bulk resistivity. If the film is not very thin, say thicker than 10 nm, it has polycrystalline structure. The grain boundaries contribute to the electron

transport (for the references see¹⁴). In a recent paper¹⁴ the optical characteristics of the films were treated as the grains with bulk material properties plus electron scattering on the grain boundaries.

The gold films with the grain size from 15 to 45 nm were deposited on glass with two different methods¹⁴. Reflectance of the films was investigated in the wavelength range 0.3-50 μm . The grain boundaries were modelled with the delta potentials, scattering from phonons and point defects were accounted by the relaxation time. The grain size distribution had no significant influence, so the films were characterized only by the mean grain size D . The material is described by the two dielectric functions longitudinal ε_l and transverse ε_t . Only ε_t changes because the grain boundaries are perpendicular to the film surface. In the local limit for ε_t the following expression was found

$$\varepsilon_t(\omega) = \varepsilon(\omega) + \frac{3\omega_p^2}{\omega(\omega + i\omega_\tau)} \left[\frac{1}{2}\alpha - \alpha^2 + \alpha^3 \ln(1 + \alpha^{-1}) \right], \quad (1)$$

$$\alpha \equiv \frac{v_F}{D\omega_\tau} \frac{\mathcal{R}}{1 - \mathcal{R}} \left(1 - i\frac{\omega}{\omega_\tau} \right)^{-1}, \quad (2)$$

where $\varepsilon(\omega) = \varepsilon_l(\omega)$ is the dielectric function of the crystalline gold, ω_τ is the Drude relaxation frequency, v_F is the Fermi velocity, \mathcal{R} is the reflection coefficient of electrons on the grain boundary. The latter is the only empirical parameter, which was found in the same experiment to be $\mathcal{R} \approx 0.65$. The reflectance of the film was expressed via the surface impedance. Comparison with the experimental reflectance showed that the model describes well the effect of the grain size except for the very small grains $D \approx 15$ nm when the electron mean free path becomes comparable with D .

We used the expression (1) at imaginary frequencies $\omega = i\zeta$ and calculated the impedances for s and p polarizations following the Kliewer and Fuchs procedure¹⁵. The impedances are different due to the nonzero wave vector \mathbf{q} along the plates in the Lifshitz formula. The details will be reported later. The relative correction to the Casimir force as a function of the grain size is presented in Fig. 1(b). The typical grain size for the films 100 – 200 nm thick deposited on the substrate at room temperature is less than 50 nm¹⁶ but it can depend on the deposition details and we take as the upper limit $D < 100$ nm.

4 Patch potential

The importance of the patch potential for the Casimir force measurement was stressed in a recent paper¹⁷. For a crystal the work function has different values for different crystallographic planes. This difference should be compensated by the potential distribution around the crystal. The deposited films are polycrystalline and for this reason there will be local variation of the

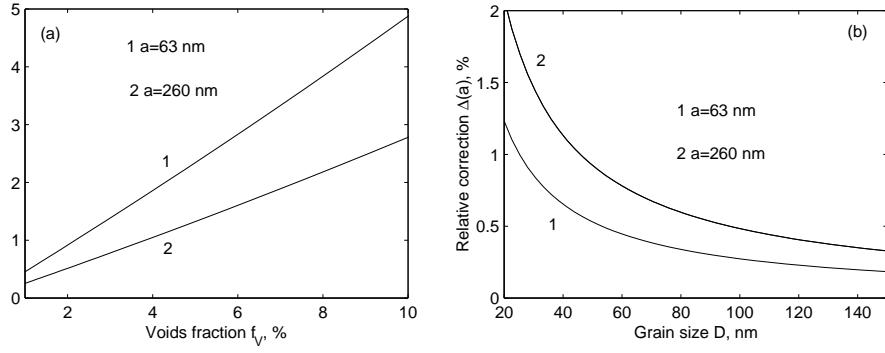


Figure 1. The relative corrections to the Casimir force $\Delta(a)$ in percents for the minimal separations explored in the experiments $a = 63 \text{ nm}$ ³ and $a = 260 \text{ nm}$ ⁵: (a) correction due to voids in the film; (b) correction due to scattering on the grain boundaries as a function of the mean grain size D

potential nearby the surface. It will result in an additional force, electrostatic in nature, which will be measured together with the Casimir force. A simple model was proposed¹⁷ to estimate the effect. In this model the dipoles were distributed on the planes. Their interaction gives the force which is defined by the spectral density of the surface potential correlation function $C(k)$. The model is in agreement with the independent *ab initio* calculation of the potential distribution near *Al* crystallite¹⁸. For the sphere and plate the force was written in the form

$$F_{patch}(a) = -\varepsilon_0 R \int_0^\infty dk k^2 \frac{C(k) e^{-ka}}{\sinh ka}, \quad (3)$$

$$C(k) = \int d^2 \mathbf{r} e^{-i\mathbf{k}\mathbf{r}} c(\mathbf{r}), \quad c(\mathbf{r}) = \langle v(\mathbf{r}) v(0) \rangle, \quad (4)$$

where ε_0 is the permittivity of vacuum, R is the sphere radius, $v(\mathbf{r})$ is the local potential on one of the surfaces and the potential distribution on different surfaces was assumed to be uncorrelated. The spectral density $C(k)$ is crucial for the force estimate. To our opinion, in the original paper¹⁷ it was overestimated. There was no clear idea around which value of k the spectral density should be centered and what is the width of this function, so these values were chosen quite arbitrary. Looking at the films surface images one can easily guess that the spectrum should be centered around the mean grain size D with the wavelengths presented roughly from $D/2$ to $2D$. If we consider the spectrum in this range as flat, the force will be the following

$$F_{patch}(a) \approx -\frac{4\pi\epsilon_0\sigma_v^2 R}{15a} \exp\left(-\frac{2\pi a}{D}\right). \quad (5)$$

Here σ_v^2 is the variance of the potential distribution that was estimated¹⁷ as $(90\text{ mV})^2$. The relative contribution of the patch potential to the measured Casimir force is estimated to be smaller than 0.4 % for the minimal separation $a = 63\text{ nm}$ in the AFM experiment³ and the largest grain size $D = 100\text{ nm}$. It decreases fast for smaller grain size or for larger separations. However, it should be stressed that the patch force mimics the Yukawa interaction and can be easily confused with a new force.

5 Discussion

Above it was demonstrated that the optical properties of deposited gold films differ from those of the bulk material. The main reasons for the deviations are the voids in the films and electron scattering on the grain boundaries. Both effects diminish the absolute value of the predicted Casimir force on the one percent level. We define the relative correction as

$$\Delta(a) \equiv \left| \frac{F_f(a) - F_b(a)}{F_b(a)} \right|, \quad (6)$$

where the indexes b and f refer to the bulk and film materials, respectively. For the AFM experiment³ in the sphere-plate geometry the expected correction was found for the most probable values $f_V = 4\%$ and $D = 50\text{ nm}$: $\Delta(63\text{ nm}) = (1.9 + 0.5)\% = 2.4\%$. The same correction for the MEMS experiment⁵ with the effective plate-plate geometry is $\Delta(260\text{ nm}) = (1.0 + 0.9)\% = 1.9\%$. One can see that these corrections cannot be ignored since in both experiments the precision was better than 1 %.

There is an additional effect which also reduces the absolute value of the predicted force. At frequencies larger than ω_p the charge density fluctuation (plasmons) can propagate in the material decreasing the reflection coefficient for p -polarization. Importance of this nonlocal effect for the Casimir force at separations of the order or smaller than plasma wavelength λ_p was stressed for the first time in the paper¹⁹ where the correction due to plasmon excitation was found. For the AFM experiment³ it is estimated as 3 % at $a = 63\text{ nm}$. Due to larger separation this correction is smaller for the MEMS experiment⁵, where it is 1 % at $a = 260\text{ nm}$. The plasmon correction was dismissed in Ref. 5 on the basis that the separation was much larger than the penetration depth $\delta = \lambda_p/2\pi$. In this connection we should note that the separation a has to be compared with λ_p but not δ . Otherwise on the same basis one could conclude

that the finite conductivity correction is negligible at $a \sim \lambda_p$ while in reality it is of about of 50% (see Fig. 1 in the paper⁷ where the transition point at $a \sim \lambda_p$ not at $\lambda_p/2\pi$ is clearly seen).

In conclusion, we demonstrated that it is important to take into consideration the optical characteristics of the metallic films used in the experiments and showed the way how the deviation from the bulk metal properties can be estimated. Voids in the films, scattering on the grain boundaries so as non-local effects all of them tend to reduce the force in comparison with the bulk metal prediction. Even without nonlocal effects the reduction is expected on the level of 2 %.

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